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ABSTRACT. In order to investigate the problem
of the effect of an electrical field on gas
adsorption, a flow method was worked out, in
which the change in the concentration in a
gaseous mixture could be determined using a
method of thermal conductivity. Increased
adsorption of carbon dioxide and sulfur di-
oxide was observed on aluminum plates with an
electrical field of 17,000 V/cm between them.
The reasons for this effect were discussed.

1. Introduction. Several years ago, N. Stark and myself /369*
[1] theoretically investigated the problem of the magnitude of
gas adsorption in an electrical field in this journal, and found
that the increase in the adsorption of a dipole gas in an electrical
field, which must be calculated using the electrostatic theory of
adsorption, is very small, if we use those fields, which would come
into account for an experimental investigation [2, 3, 4]. A cal-
culation of the fields on ion lattice surfaces yielded field
strengths, which indicated the probability of an electrical ad-
sorption on these crystal surfaces.

According to our assumption, the orientation of the dipole
molecule in the field of its reflected, electrostatic images should,
as the Lorenz-Landé electrostatic theory of adsorption [5] assumes,
be intensified by an external electrical field, and thus result in
an increase in the adsorption. The fact that there is no appreciable
difference in the adsorption of a dipole gas, such as sulfur dioxide,
or a quadrupole gas, such as carbon dioxide, contradicts the electro-
static theory of adsorption. However, the electrostatic theory of
the van der Waals forces of attraction of Keesom (quadrupole forces)
and Debye (dipole forces) was not satisfactory. However, the

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* Numbers in the margins indicate pagination in the foreign text.

mechanical wave theory led to the view that the forces of
attraction can be explained if the charge distribution is
assumed to be statistical when there is a centrally symmetrical
charge distribution in the molecule, and that the electron
lead to a polarization which generates a constant attraction.
London showed more precisely that the non-static charge distribu-
tion in the molecules (electron motion) produces mutual forces of
attraction, which can occur both as van der Waals forces as well
as adsorption forces, the identity of which was also supported by
Eucken. However, we can expect no theoretical basis for our problem
from this, which would lead to the assumption of a greater effect
than is the case using the electrostatic theory, which, for all
practical purposes, can be considered valid along with the mechanical
wave theory. A new assumption presented itself only insofar as the
adsorption is caused by the effect of the wave mechanical forces of
adsorption, and the electrostatic forces (**with and** without the
supplementary electrical field) and exhibit their effects in the
adsorption layer [6]. In this layer, it cannot be expected that
we are still talking about a gaseous state of the molecule, and we
can use the dielectric saturation phenomena and other phenomena for
purposes of comparison. Finally, there are electrical charge phe-
nomena in the case of gas adsorption, which we would expect to be
influenced by an electrical field.

2. Measurement Technique. The general methods of measuring
adsorption [2] involve determining the strength of the adsorption
from the absorption of the gas, i.e. a pressure measurement, or
determining the thermal effect of the adsorption. The second method
was not sensitive enough for the purpose at hand, i.e. measuring the
change in the adsorption under the effect of an electrical field
and comparing it with the one without the field. It was also im-
possible to carry out the measurements using pressure measurements
because it was feared that, because of the high sensitivity of the
pressure measurement, we would observe a pressure change caused
caused by the electrostriction of the gas in addition to or instead
of a pressure change caused by increased adsorption. The best and

most reliable method would be the use of the Mc Bain adsorption scale, but these scales are only constructed with the required sensitivity with very small load capacities[1], whereas we would be working with large metal surfaces, and thus an adsorbent volume of considerable weight. The last method would be a flow method   /371 for gas mixtures, which has already been used several times for normal adsorption measurements. At my request, Mr. V. Schwab the apparatus, described in the following paragraphs, and used it to carry out a series of measurements.

The gases, which were investigated, were carbon dioxide and sulfur dioxide; they were supposed to be adsorped on metallic aluminum, which formed the coat of a multiple plate condenser, to which an electrical field could be applied. The gas, which was to be studied, was mixed with hydrogen in a glass globe G_1 (6 liters) (usually at a ratio of 1 : 1). The mixture was then allowed to flow, choked by capillary tubes of the appropriate diameter, through the adsoprtion condenser K into a second, completely evacuated glass globe G_2 (3 liters), under the effect of its own pressure.
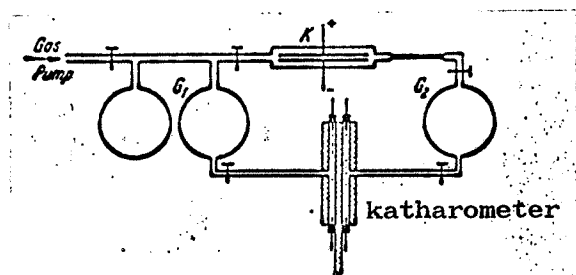

Figure 1

As soon as the pressure in the first globe dropped to a specific level, the globes were sealed off from each other by a stopcock, and the gas mixtures in G_1 and G_2 subjected to a relative gas analysis using the thermal conductivity method. The schematic structure of the apparatus is shown in Figure 1.

The carbon dioxide and the hydrogen were generated in Kipp generators and carefully dried. The sulfur dioxide was obtained from a bomb. It was absolutely anhydrous. A manometer was used to produce the gas mixture. The initial pressure of the mixture was usually slightly less than the external barometric level.

1. The effects of electrostatic forces would also be difficult to avoid.

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In order to obtain a better mixture, the mixing globe was
heated on the side with a hot-air blower. Usually the gas mixture
stayed in globe G_1 more than 12 hours. The actual adsorption
vessel consisted of a hard glass tube, 60 cm long and 4.1 cm in
diameter (Figure 2), which housed the adsorption condenser. The
condenser itself consisted of 300 small aluminum plates with a
total surface area of 11 cm^2, which were ordered with spacers so
that the interval between them was less than 1 mm (Figure 2a).
The base and head were made of marble plates which were connected
by four brass pins. Adjacent aluminum plates were each in contact
with two of these brass pins. The aluminum sheet was 0.3 mm thick.
After the small plates were cut out, they were polished to a high /37
sheen and degreased. The plates stayed highly polished right until
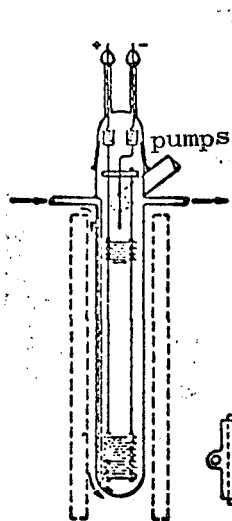the end of the experiments. However, it must be assumed that a
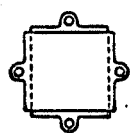thin layer of the oxide will never-
theless be deposited on the surface.
An electric oven could be inserted
above the adsoprtion chamber, with
which the chamber and the condenser
could be heated to more than 400°C.
By simultaneously removing the libera-
ted gas radicals using a mercury dif-
fusion pump as a high-vacuum pump and
a Cenko pump as a preliminary pump,
the condenser was completely degassed.
The Cenko pump also served to evacuate

Figure 2   Figure 2a

globes G_1 and G_2. The gas mixture
entered the (cooled) adsorption chamber through a thin glass tube,
and then flowed through the condenser upward and through the capil-
lary tubes to the globe G_2. The flow process usually took ½ hour
so that we could assume a careful adjustment of the adsoprtion equi-
librium in the case of adsorption on blank surfaces. The capillary
tubes were mounted to the rear of the adsorption chamber, so that
thay were under high pressure from the start of the experiment. It
was assumed that the heavy gas would be adsorbed when the gas mixture

4.

flowed through the condenser to the metallic surface, since hydrogen is only weakly adsorbed by aluminum surfaces (according to Sieverts). The change in the gas mixture should be determined by analysis, once in a test without an electric field at the condenser, and once under the same conditions, but with an electrical field. In preliminary tests, the dissociation through adsorption on the glass walls of the adsorption chamber (without condenser) was determined.

The analysis of the gas mixture was carried out relatively using a thermal conductivity method. The thermal conduction chamber (katharometer) consisted of a brass cylinder with a diameter of 6 cm and a length of 18 cm (Figure 1). Two holes were bored into it, through which the brass tubes, with a diameter of 0.4 cm, could be inserted. These brass tubes, which had good thermal contact with the brass block, contained a platinum wire, with a diameter of 0.2 mm down the center, the heating wire, which was constantly kept taut by small steel springs on the side. /373

The two platinum heating wires formed the two arms of a sensitive Wheatstone bridge with a 1 m long measurement wire, which was sensitized on both sides by applying resistances of 100 ohms. In order to determine the zero voltage in the bridge, a pointer galvanometer was used for the rough determination and a mirror galvanometer which could be switched. A current of 1 ampere was supplied by the current source during the measurement. This current was controlled with a mirror galvanometer with reflecting scale. The strength of the current was kept constant by compensation, which proved to be unnecessary in view of the magnitude of the observed changes in the resistance.

The principle of the method of thermal conductivity is based on the known fact that the temperature of the wire, which a steady heating current flows through, is a function of the thermal conductivity of the surrounding gas, but the temperature, on the other hand, can be measured by the electrical conductivity of the wire. If the wire is in a gaseous medium, the components of which have a

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varying thermal conductive capacity, then a change in the make-up of the mixture will also induce a change in the thermal conductivity, which will be easier to observe, the greater the difference between thermal conductivities of the pure gases. Hydrogen has a relatively high thermal conductivity and thus lends itself well for use as a mixing gas. The katharometer must be calibrated for the appropriate pairs of gases through preliminary tests.

Glass tubes are used to connect the differential katharometer to the glass globes. The katharometer tanks were evacuated. By opening the stopcocks, the gas flows from the globes into the katharometer chambers. Pressure differences, which in this case are very small, play no role because the thermal conductivity in these pressure ranges is completely independent of the pressure. If the gas mixtures from G_1 and G_2 were not of the same composition, this would be indicated by a deflection of the galvanometer pointer in the bridge. In order to compensate for this on the basis of the calibration, a measurement for the type and magnitude of the change in composition was used. In order to control the calibration, a small amount of hydrogen was added during each test to the mixture from G_1, which thus remained unchanged, and the change in the thermal conductivity which resulted was observed. In the preliminary tests, it was also established whether a change in the composition of the mixture occur while its was standing in the globe. These observations ran negative. The globes were maintained completely free /374 of water, since a solution of the very soluble gases CO_2 and especially SO_2 could have produced a simulation of the adsorption effect, although this would not have been the case with the pure adsorption.

The high-voltage source for the voltage of the adsorption condenser consisted of a 120/8,000 V transformer, which included a potentiometer on its primary side. The secondary voltage was rectified by a hot-voltage rectifier, stabilized by a capacitance and measured with a Braun electrometer. The voltage was supplied to the adsorption chamber through brass pins, which were cemented

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The magnitude of the applied voltage was limited by spark dis-
charges at the condenser, which first occurred at points. In order
to definitely avoid discharges, the voltage was not raised above
2,000 V. Disruptive discharges could be noticed immediately by
a voltage breakdown at the Braun electrometer. Occasionally, an
insulation check was made by inserting a galvanometer.

3. Measurement Results. The adsorption tests were repeated
several times with mixtures of the same composition, both without
and with an applied electrical field of varying field strength.
Since the average distance between the plates in the absorption
condenser was 0.9 mm, the voltages applied to the plates must be
multiplied approximately by a factor of 11 in order to obtain the
values of the electrical field strength in [V/cm].

The adsorption tests with carbon dioxide were carried out at
room temperature and -80°C. In this case, a large double-walled
metal chamber, which was always connected to the Cenko pump, was
filled with a paste of CO_2 in toluol and inserted above the adsorp-
tion chamber. In the tests with sulfur dioxide, the same double-
shell vessel was filled with a cooling mixture, which brought the
temperature of the adsorption chamber to -10°C. The following
Tables 1 and 2 contain the results of the measurements. The same
method was consistently used to degas the adsorption chamber between
the tests. The sequence of the data corresponds to that of the
measurements. Preliminary tests are not listed.

We notice immediately that the tests, in which the final
pressure was higher, i.e. the flow was interrupted at an earlier
point, exhibit sharper changes in the mixture, i.e. increased ad-
sorption. Since an increase in the hydrogen content of the mixture
in G_2 was observed in all cases, this at least means that there was
a heavier adsorption of CO_2 on the aluminum. Since it could   /375
not be determined from our apparatus how large the absolute
amount of the adsorbed gas actually was, the results of the experi-
ment naturally do not preclude the possibility that some hydrogen

TABLE 1. ADSORPTION OF CARBON DIOXIDE

Final pressure in cm Hg	Voltage at ads. cond. in volts	Temperature, °C	% hydrogen in mixture in G_1	Increase in % of hydrogen in G_2
37	1,000	24	40.4	0.5 ± 0.05
25.3	1,200	25	52.3	0.6
29.2	0	18	51.4	0.12
29	900	18.5	50.7	0.13
28	900	21	50.9	0.12
29	900	19.5	50.8	0.13
35.1	0	19	52.9	0.32
27.6	0	−80	51.8	0.19
28.3	900	−80	51.3	0.19
50.5	1,500	24	50.3	1.52
49.9	0	26	52.8	0.43
48.7	1,300	22	51.32	1.12
47.5	1,500	−80	50	0.17
51.7	1,500	25.2	50	1.2
52.1	1,500	25.2	50.1	1.2
50.9	1,700	20	50	1.8
50.3	1,700	−80	49.7	0.42
50	1,000	−80	50.0	0.21
49.9 w/o heat	0	−80	50.1	0.16
47 w/o cond.	0	22	50	—
51.2	0	23.1	51	0.08

TABLE 2. ADSORPTION OF SULFUR DIOXIDE

Final pressure in cm Hg	Voltage at ads. cond. in volts	Temperature, °C	% hydrogen in mixture in G_1	Increase in % of hydrogen in G_2
50.2	1,500	24	50.5	0.46 ± 0.05
52.5	1,500	25	49.5	0.5
49.4	0	24.6	49.7	0.27
49.3	0	21.2	49.6	0.27
51.3	1,000	26	50.3	0.3
49.7	1,800	23	50	1.0
49.5	2,000	24.5	49.8	1.37
49.1	2,000	21	49.1	1.31
50.8	2,000	−10	48.9	1.49
50.1	0	−10	50.2	0.5
52.5	1,000	−10	48.6	0.47
50.4	1,000	−10	50.3	0.66
50.3	1,500	−10	50.7	0.87
50.5	1,800	−10	50.2	1.1

TABLE 2. [CONT.]

Final pressure in cm Hg	Voltage at ads. cond. in volts	Tempera-ture, °C	% hydrogen in mixture in G_1	Increase in % of hydrogen in G_2
w/o heat 53.2	0	21.8	50	0.10
w/o cond. 50.1	0	-10	50	0.17

adsorbs at the aluminum. However, we can observe a increase <u>/</u>376
in the percentual dissociation in favor of the adsorbed carbon
dioxide in the case of an electrical field in the adsorption
chamber. Figure 3 shows the change in the mixing ratio of the gas
composition as a function of the applied voltage, both for room
temperature (18 to 25°C) and for -80°C. From the values in Table 1
for the percentual change, we subtract the same value without the
condenser, and it is these figures which are used in the text.

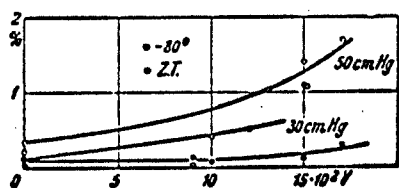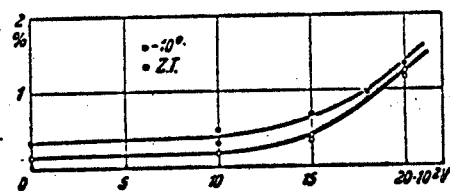In the same way, the tests were carried out for the sulfur
dioxide, which was also mixed with
hydrogen. In this case, in view of
the results of the tests with CO_2,
the experiment was interrupted when
the final pressure was higher, and
we were able to work with slightly
higher voltages at the adsorption
condenser. The results are given in
Table 2 and Figure 4.

Figure 3

Figure 4

The results of the adsorption
test with sulfur dioxide show that
even in this case, the observed per-
centual increase in hydrogen in the
gas mixture is the reason why the ad-
sorption of sulfur dioxide on the aluminum was heavier. In this
case, in contrast with the results for carbon dioxide, there is

an increase in adsorption of SO_2 at lower temperatures, as is expected, i.e. no special influence on the part of the adsorbed hydrogen. This is obviously connected with the fact that hydrogen at -80°C, as in the CO_2 tests, is adsorbed more readily than in the tests with SO_2 which were carried out at only -10°C, and, as a result, the probably also heavy adsorption of CO_2 at -80°C cannot be observed in our experiments.

The general nature of the curve, however, does not correspond to any linearity between the increase in adsorption and the field strength. The dissociation caused by adsorption is approximately the same for both mixtures, so that, assuming that the adsorption of hydrogen from both mixtures is the same at room temperature, the adsorptions of CO_2 and SO_2 are also almost the same and are /377 influenced to the same extent by the electrical field. However, the effect of the fact that the one gas is dipolar (SO_2) is not of any great importance for the experiment. For the following discussion of the test results, let us assume that the adsorption of hydrogen at room temperature is negligiable with respect to the adsorption of the other gases.

Whether the adsorption is "polar", i.e. only heavy at the positive or negative plates, cannot be determined. In tests dealing with this specific problem, the plates would have to be mounted against thin wire screens, i.e. the adsorption surface of a different potential would have to be a different size. Such a case would be expected for a dipole gas, the dipole of which is situated asymmetrically in the molecule [7].

The somewhat stronger effect of the electrical field on the adsorption of carbon dioxide relative to that of sulfur dioxide is possibly only an apparent one, and could be connected with the adsorption of hydrogen. For a continuation of the experiments, it would be important to figure out the true amounts adsorbed. However, for the large volumes, which are being used, a very exact pressure measurement would have to be used at high pressures. As

a control for the values calculated below for the amounts adsorbed, the gas was eliminated through heating after an adsorption test; it was not pumped out but rather fed into a three-liter globe (G_2), which had previously been completely evacuated, and the gas pressure in this globe measured with a Mc Leod manometer. The same order of magnitude as above was obatined for the amount adsorbed. This method is too inaccurate for a more exact determination.

4. Discussion of the Measurement Results. Before proceeding any further in explaining the effect of the electrical field, the thickness of the adsorption layer must be calculated for the case of adsorption without a field. Each aluminum plate had a geometrically effective area of 11 cm^2 on both sides. Thus, the total adsorption surface for the 300 plates was 3,300 cm^2. If we assume that the surface covered by an adsorbed molecule equals $1 \cdot 10^{-15} cm^2$, then there would be $3.3 \cdot 10^{18}$ molecules in a layer one molecule thick and covering the entire geometric surface. If we now wish to find the number of molecules which are trapped while flowing through the adsorption condenser, we might obtain an estimate in the following manner. The total amount of gas in the mixture was 6 liters, of which three liters was almost always hydrogen, and three /378 liters the second gas (CO_2 or SO_2). The pressure of the mixture in globe G_1 was approximately 70 cm Hg in the experiments. By connecting the mixing globe with the adsorption chamber, which together with the feed lines had a volume of approximately 1 liter, the pressure immediately dropped to approximately 60 cm Hg, corresponding to an increase in the volume from 6 to 7 liters. From that point on, the pressure continued to drop slowly until it reached 50 cm Hg, at which point the experiment was halted. Of the existing 7 liters of the gas mixture at the beginning of the experiment at 60 cm Hg pressure, 8.4 liters were measured at the end of the experiment at 50 cm Hg pressure so that therefore 1.4 liters escaped through the capillary tubes. Half of this amount was either CO_2 or SO_2, i.e. 0.7 1 at 50 cm Hg pressure, or, converted

to atmospheric pressure, 0.46 1 = 460 cm^3. Since for field 0, the percentual dissociation after flowing through the adsorption condenser was approximately 0.2%, 0.9 cm^3 CO_2 (SO_2) would have been adsorbed, all under the assumption that the adsorption of hydrogen at room temperature is negligiable, and that the observed dissociation is the result only of the adsorption of the CO_2 or SO_2. However, there are 2.4 · 10^{19} molecules in 0.9 cm^3 of a gas at 1 atm. If we compare this number with the number for the molecules contained on the geometric surface in a monomolecular layer, we obtain a factor of 7. This factor can easily be explained by the fact that the true adsorption surface is probably 10 to 100 times larger than the geometrically determined one, so that for our experiments, we cannot assume a complete coverage of the total, actual adsorption surface.

The experiments in the electrical field show that there is an increase in adsorption, which for the two gases in question is approximately 1% when the voltage is 1,500 V, i.e. when the field is approximately 17,000 [V/cm]. The amount of gas adsorbed in this field is thus approximately five times as large as that adsorbed without the field (factor 35). Even this amount of gas can be distributed in a monomolecular layer on the true adsorption surface in accordance with what has already been said, without having to assume polymolecular layers. Obviously this does not preclude the possibility that adsorption occurs mainly at the points and edges, and that, especially when the field is applied at points where the field strength is high, a type of "liquification" will occur in the form of a "puddle formation". Studies carried out at higher pressures and correspondingly higher field strengths with /379 longer periods of flow or smaller adsorption surfaces could possibly provide information on other conditions, especially the formation of polymolecular layers in an electrical field.

As has already been mentioned in the introduction, it is not easy, in light of contemporary, theoretical notions on the forces of adsorption, to determine what effect the electrical field should have, or whether this effect would have to be expected only at

field strengths, which are much higher than the ones used in this case [7]. The explanation, which seems possible, lies in the direction already indicated above. The unspecific adsorptive effect on the dipole and quadrupole gas shows that the adsorption occurs as a result of exchange forces. They are the reason why adsorption occurs at all, i.e. that the molecule is attracted and trapped at the wall of the adsorbent at distances of an order of magnitude of 10^{-8} cm. In these ranges, however, the electrostatic forces of reflection are considerable (in comparison to kT) and the directional forces of an external electrical field are almost comparable. Even the dipoleless molecule has an induced moment. Although we previously attempted to explain the increase in adsorption through an electrical field in the sense of the electrostatic theory by the fact that the adsorption energy through the field exceeds the thermal energy of the gas molecules at greater distances from the wall, this is no longer necessary in view of the new interpretation of the process of adsorption. The external, electrical field can thus influence the molecules which are brought into contact with the wall and in the sense of the dynamic interpretation of the process of adsorption by the fact that it reduces mobility in the adsorption layer, or increases the duration. The effect of the electrical field within the adsorption chamber can also be seen in the fact that the re-evaporation of the adsorbed molecule occurs preferentially after the adsorption chamber even at low angles. This interpretation can be supported by experiments in order to measure the durations even in apparatuses, which can be charged to high voltages.

The Oliphant method [8] also seems suited to pursue the problem further. This method is related to the one which we used, but was not made known until after our tests had been completed. In this method, the liquid adsorptive is moved in a static gas /380 mixture. The falling drops of Hg produce a concentration in the lower portion of the mixture, which is caused by the adsorption of the one gas (CO_2), in which case, the droplet should   again

give off the adsorbed gas by joining together with other droplets,
and this partial dissociation is observed interferometrically.
It would be possible to give the droplets high charges with dif-
ferent charge directions. It would also be possible to transfer
them to solid adsorbents by dropping a fine powder, which could
also be charged. However, in the case of powders, we would have
to remember that the surface could only be roughly determined,
but this is not very important if the similarity between all the
tests is guaranteed.

Another explanation of the results would be that only certain
active spots adsorb on the surfaces, and at these spots the adsorbent
has the characteristics of a liquid. The electrical field would
thus influence the liquid, i.e. the molecules with strong mutual
attraction. In these liquids, the known saturation phenomena would
occur as soon as an electrical or magnetic field is applied, which
would result in a reduction of the dielectric constants etc. The
observed increases in the viscosity of liquids and solutions in an
electrical field, which are much greater than would have been theo-
retically expected, are also connected with this [9, 10, 11].

Perhaps in this case a role is played by the electrical effects
in the electric influence observed in this case on adsorption, since
they can occur during adsorption. Thus, for example, gold can be
given a charge in hydrogen or oxygen. According to Mc Bain, a
certain percentage of ions is produced from the adsorbed gas mole-
cules by the addition of freeelectrons, which can in part enter
the gas chamber with their charge. Obviously these ions can be
influenced by the field.

The effect of an electrical field on the adsorption process
was examined under controlled conditions for the first time in
the preceding experiments. It seems possible that, even in the
case of the well-known clean-up phenomena during gas discharges,
which occur at low pressures, the gas absorption is connected
partially with an adsorption of the molecules in the electrical /381

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field, and not only with the effect of ions. The electrical
fields in x-ray tubes are also substantial. On the other hand,
we cannot assume that our tests with clean-up phenomena in the
adsorption condenser are related. First of all, the tests were
carried out at relatively high pressures, which were far below
the breakdown field strength; spray discharges did not occur at the
voltages used. Secondly, the effect would not have been a function
of the temperature to the same extent, and thus it would have to
be expected that it is a function of the pressure.

No practical use can be anticipated for the electrically
increased adsorption because the technically used adsorbents are
used in the form of finely distributed powder. It would be easier
to conceive of a practical importance for the increased condensation
phenomena which are observed in the electrical field. Tests will
be carried out in this direction.

Prague. Physics Institute of the German University, June 1937

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